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A spacially extended system of two-level atoms interacting with a resonant electromagnetic field exhibits conditions for which two phases, of high and low excitation, coexist spacially, in the direction of propagation in the material. The spacial first-order phase transition and its properties are related to an inversion-dependent renormalization of the resonance frequency, which becomes significant for a collection of two-level atoms with high density and large oscillator strengths.

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LONGITUDINAL SPACIAL FIRST-ORDER PHASE TRANSITION IN A SYSTEM OF COHERENTLY-DRIVEN, TWO-LEVEL ATOMS

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A spacially extended system of two-level atoms interacting with a resonant electromagnetic field exhibits conditions for which two phases, of high and low excitation, coexist spacially, in the direction of propagation in the material. The spacial first-order phase transition and its properties are related to an inversion-dependent renormalization of the resonance frequency, which becomes significant for a collection of two-level atoms with high density and large oscillator strengths.

The phenomenon of optical bistability (OB) is usually based upon two components: (a) The illumination of an optically nonlinear material; (b) A feedback mechanism which is provided by the mirrors of a high-Q cavity [1]. It was first shown by Bowden and Sung [2] that OB without external feedback may occur for a system composed of a collection of two-level atoms interacting via the electromagnetic field and driven by an externally applied coherent field. By using a fully quantized many-body model in the mean-field limit, they have shown [2] that the role of the feedback may be provided intrinsically by the induced coherent dipole–dipole interactions among the atoms of the material.

For sufficiently low values of the incident field, the reaction field produced by the induced dipoles opposes the externally applied field and thus reduces the net field. For sufficiently large values of the incident field, the individual atom's interaction with the electromagnetic field is influenced much more strongly by the externally applied field than the reaction field due to the interatomic interaction. The incident field overwhelms the interatomic interaction via the electromagnetic field, and thus the contribution of the reaction field due to the coherent dipole–dipole interactions is largely negated. This occurs as a first-order phase transition far from thermodynamic equilibrium.

Recent interest in intrinsic optical bistability (IOB) has stimulated a surge in theoretical and experimental work [3–13]. Particularly relevant to the work presented here is the recent theoretical discussion of the effect of the insertion of the local-field correction in the usual Maxwell-Bloch formulation in causing IOB [12], and the origin of the need for that correction in the passage from microscopic to macroscopic electrodynamics [13] in semiclassical theory. The origin of IOB was identified in that treatment as due to an inversion-dependent renormalization of the atomic transition frequency caused by coherent dipole–dipole interaction among two-level atoms. Previous quantum mechanical treatments of the problem [2] used a mean-field approximation in a volume much smaller than a cubic wavelength and suffered from the defect that propagation and retardation effects were neglected. Although the treatments using semiclassical theory include these effects within the framework of the semiclassical approximation, the recent work was been addressed to either a propagation length much smaller than a resonance wavelength [12] or to a volume containing a specified (small) number of atoms [13].

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In more recent works [14–16] we consider the problem from a many-body quantum mechanical approach in which we treated a large number of driven, interacting, spacially distributed two-level atoms. The intrinsic many-body statistics justify factorization in the multiplication of the dipole operators [15], while propagation and retardation effects are included and found to be important [14,15]. It has been shown [14,15] that interatomic correlation is built up in the direction of propagation of the incident field and, that under appropriate conditions and limits, the usual Maxwell-Bloch equations are reproduced. However, for high densities and large oscillator strengths, frequency renormalization which is inversion-dependent appears as a modification to the usual Bloch equation for the complex atomic dipole operator [14–16].

In this letter we show that, due to the nonlinear frequency-renormalization correction, a new kind of spacially dependent first-order phase transition may occur in the interaction between a coherently driven, interacting collection of spacially distributed two-level atoms of large density and high oscillator strengths. We show how the two phases of high and low excitation may coexist spacially in the propagation direction in the material. Due, in part, to this longitudinal spacial dependence, the first-order phase transition predicted here for IOB is fundamentally different from the well-known effect of bistability in resonators.

The equations of motion derived by us are given as [14,15]

$$d\langle\sigma_z\rangle/dt = -\gamma_L[\langle\sigma_z\rangle + 1] + (\mu/\hbar)[E^*\langle\sigma_{+0}\rangle + E\langle\sigma_{-0}\rangle], \qquad (1)$$

$$d\langle\sigma_{+0}\rangle/dt = -(\mu E/2\hbar)\langle\sigma_z\rangle + i[\Delta - \epsilon\langle\sigma_z\rangle]\langle\sigma_{+0}\rangle - \gamma_T\langle\sigma_{+0}\rangle, \qquad (2)$$

$$(1/c)\partial E/\partial z = -(4\pi\omega n/c)\langle\sigma_{+0}\rangle. \qquad (3)$$

Here, E is the slowly-varying envelope of the electric-field amplitude, $\langle\sigma_{+0}\rangle$ is the expectation value of the slowly-varying complex atomic polarization per unit volume, and $\langle\sigma_z\rangle$ is the expectation value of the atomic inversion per unit volume. The equations for E^* and $\langle\sigma_{-0}\rangle$ are the complex conjugates of eqs. (3) and (2), respectively. The parameter μ is the modulus of the matrix element of the transition dipole moment of an atom, ω is the frequency of the incident field and n is the number of atoms per unit volume. The rates γ_L and γ_T are the inverses of the relaxation times T_1 and T_2 for the inversion of population and the dipole moment, respectively, and $\Delta = \omega - \omega_0$ is the deviation of the applied field frequency ω from the atomic resonance frequency ω_0. Finally, ϵ is the frequency renormalization constant derived in our previous work [14,15]

$$\epsilon = 7\pi n\beta c^3/4\omega_0^3, \qquad (4)$$

where β is the spontaneous decay constant

$$\beta = 4|\mu|^2\omega_0^3/3\hbar c^3. \qquad (5)$$

The frequency renormalization stems from coherent dipole–dipole interactions between atoms that are within a volume of a cubic wavelength [2,12–15].

In the fully quantum mechanical model, $\gamma_L = \beta$, $\gamma_T = \beta/2$, but by considering additional homogeneous broadening, we may treat γ_L and γ_T as empirical constants. The usual Maxwell-Bloch equations [17] are obtained from eqs. (1)–(3) in the limit $\epsilon \to 0$. The frequency renormalization correction proportional to ϵ becomes important only for a two-level system with high oscillator strengths and a large density. Such a system can be realized in experiments with Rydberg atoms. While the analysis presented here is suitable mainly for an atomic system, it suggests that similar effects may be obtained by using crystals with a high density of bound excitons having large oscillator strengths, which may be realized in CdS for bound I_2 excitons [18].

Under steady-state conditions we get, from eqs. (1)–(3):

$$\langle\sigma_{+0}\rangle = \mu E\langle\sigma_z\rangle/2\hbar[i\Delta - \gamma_T - i\epsilon\langle\sigma_z\rangle], \qquad (6)$$

$$\gamma_L[\langle\sigma_z\rangle + 1]\{\gamma_T^2 + [\Delta - \epsilon\langle\sigma_z\rangle]^2\} = -\gamma_T I\langle\sigma_z\rangle, \qquad (7)$$

$$\partial I/\partial z = -\gamma_L[1 + \langle\sigma_z\rangle], \qquad (8)$$

where I is the normalized intensity of radiation,

$$I = |\mu E/\hbar|^2, \qquad (9)$$

and

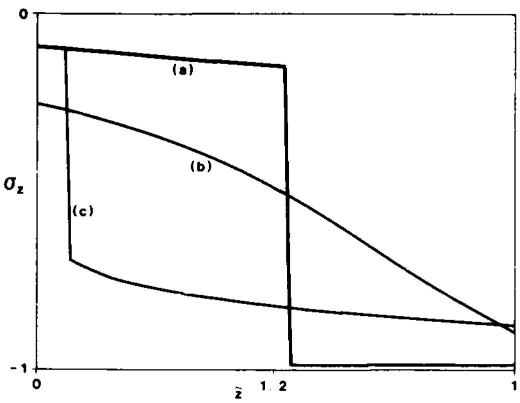

Fig. 1. The atomic inversion of population $\langle\sigma_z\rangle$ is calculated as a function of the normalized distance $\bar{z}$ [defined in units of s^{-1} by eq. (10)] using the normalized parameters: $\gamma_L=2$, $\gamma_T=1$, $\Delta=-4$ (in units of s^{-1}). The intensity of the incident radiation I [defined in units of s^{-2} by eq. (9)] is given by $I(\bar{z}=0)=100$. Curves (a) and (c) are calculated according to eqs. (7) and (8) with the parameter $\epsilon=20$ (s^{-1}) using, respectively, the upper and lower branch solution of the cubic equation. Curve (b) is calculated according to eqs. (11) and (12) with parameter $\epsilon=0$ (the regular Maxwell-Bloch equations).

$$\bar{z}=(4\pi\mu^2\omega n/\hbar c)z\,. \tag{10}$$

The normalized distance $\bar{z}$ and the constants γ_T, γ_L, Δ and ϵ are given in units of s^{-1} while I is given in units of s^{-2}.

Eq. (7) is a cubic equation leading to intrinsic local bistability because of the nonlinear term which is proportional to ϵ. Eqs. (7) and (8) can be numerically integrated to give the spacial distribution of excitation in the extended medium, given $I(z=0)$.

For the usual Maxwell-Bloch equations ($\epsilon=0$), we get simple analytical solutions:

$$\langle\sigma_z\rangle=-\gamma_L(\gamma_T^2+\Delta^2)/[\gamma_L(\gamma_T^2+\Delta^2)+\gamma_T I] \tag{11}$$

representing the usual saturable nonlinearity, and

$$\gamma_L(\gamma_T^2+\Delta^2)\ln[I(\bar{z})/I(0)]$$
$$+\gamma_T[I(\bar{z})-I(0)]=-\gamma_L\gamma_T\bar{z}\,, \tag{12}$$

which yields only one solution (no bistability) for I at each $\bar{z}$ [1].

In fig. 1 are shown results of some of our calculations for $\langle\sigma_z\rangle$ as a function of $\bar{z}$. We have taken $\gamma_L=2$ so that all rates, including $\bar{z}$, are in units of $(\gamma_L/2)$, while I is in units of $(\gamma_L/2)^2$. All the curves in fig. 1 of $\langle\sigma_z\rangle$ as a function of $\bar{z}$ have been calculated by using the parameters $\gamma_L=2$, $\gamma_T=1$, $\Delta=-4$, $I(\bar{z}=0)=100$. Curves (a) and (c) have been calculated according to eqs. (7) and (8) with the parameter $\epsilon=20$, using, respectively, the upper and lower branch solutions of the cubic equation [eq. (7)]. For comparison with the ordinary Maxwell-Bloch equations, we describe in curve (b) the calculation of I as a function of z according to eqs. (11) and (12), (i.e., for $\epsilon=0$).

By starting with a high intensity of the incident electromagnetic field and reducing its strength, we expect $\langle\sigma_z\rangle$ as a function of z to follow the upper-branch solution, of the bistable region, as demonstrated in fig. 1 by curve (a). By starting with a low intensity for the incident field and increasing its strength, we expect $\langle\sigma_z\rangle$ as a function of $\bar{z}$, for the same set of parameters, to follow the lower branch solution, as demonstrated in fig. 1 by curve (c). A spacial first-order phase transition will occur for the two cases at different distances along the long sample, for the same incident intensity. In order that the effect will be pronounced, the value of ϵ should be large (high density and large oscillator strengths). Also, in order to get the phase transition, the frequency of the coherent radiation should be lower than the atomic transition frequency and of such a value that the frequency renormalization will bring it in and out of resonance as a function of the value of $\langle\sigma_z\rangle$ (as demonstrated in fig. 1 by using the values $\Delta=-4$, $\epsilon=20$).

To obtain a good condition for intrinsic bistability, the parameter ϵ should fulfill the condition $\epsilon/\gamma_T>6$, where γ_T is the relaxation rate for the complex dipole. Assuming for the pure quantum case $\gamma_T=\beta/2$ where β is the spontaneous decay rate, we have $\epsilon/\beta>3$ and we find that we need approximately a few hundred (or more) atoms within a volume of a cubic wavelength in order that the effects will be significant. If we introduce additional homogeneous broadening, which increases the value of γ_T, the density should be increased by an additional factor $2\gamma_T/\beta$. The present effects are therefore important only for high densities and large oscillator strengths. This is the main reason why in ordinary transmissivity experiments, these effects are negligible and the usual use of the Maxwell-Bloch equations ($\epsilon\simeq0$) is justified. Also, one should note that the longitudinal spa-

cial first-order phase transition is predicted here as an internal effect in the material for the inversion of population along the sample, for certain critical parameters. The observation of this effect needs, therefore, the use of transverse probe fields by which local inversions of populations are detected.

In the present analysis we have neglected the effect of output boundary conditions, which is a quite good approximation for a very long sample. Also, we have assumed in our model a homogeneous broadening. The spacial first-order phase transition obtained in the present theoretical analysis is a fundamentally new effect in quantum optics. We predict here the possibility for its observation in experiments, where the inversion of population $\langle\sigma_z\rangle$ can be measured as a function of $\tilde{z}$ by using a probe electromagnetic field propagating in the transverse direction of a long sample.

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